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Assessing the Bioaccumulation of Contaminants from Sediments of the Upper Mississippi River Using Field-Collected Oligochaetes and Laboratory-Exposed *Lumbriculus variegatus*

E. L. Brunson, T. J. Canfield,* F. J. Dwyer, C. G. Ingersoll, N. E. Kemble

Environmental Contaminants Research Center, United States Geological Survey, 4200 New Haven Rd., Columbia, Missouri 65201, USA

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Abstract. Concern with the redistribution of contaminants associated with sediment in the upper Mississippi River (UMR) arose after the flood of 1993. This project is designed to evaluate the status of sediments in the UMR and is one article in a series designed to assess the extent of sediment contamination in navigational pools of the river. Companion articles evaluate sediment toxicity and benthic community composition in navigation pools of the river. The objectives of the present study were to: (1) to assess the bioaccumulation of sediment-associated contaminants in the UMR using laboratory exposures with the oligochaete *Lumbriculus variegatus,* and (2) to compare bioaccumulation in laboratory-exposed oligochaetes to field-collected oligochaetes. Sediment samples and native oligochaetes were collected from 23 navigational pools on the Upper Mississippi River and the Saint Croix River. Contaminant concentrations measured in the *L. variegatus* after 28-day exposures to sediment in the laboratory were compared to contaminant concentrations in field-collected oligochaetes from the 13 pools where these sediments were collected. Contaminant concentrations were relatively low in sediments and tissues from the pools evaluated. Only polycyclic aromatic hydrocarbons (PAHs) and total polychlorinated biphenyls (PCBs) were frequently measured above detection limits. The majority of the biota-sediment-accumulation factors (BSAFs) for PAHs were within a range of about 1.0 to 2.6, suggesting that the theoretical BSAF value of 1.7 could be used to predict these mean BSAFs with a reasonable degree of certainty. A positive correlation was observed between lipid-normalized concentrations of PAHs detected in laboratory-exposed and field-collected oligochaetes across all sampling locations. Rank correlations for concentrations of individual compounds between laboratory-exposed and field-collected oligochaetes were strongest for benzo(e)pyrene, perylene, benzo(b,k)fluoranthene, and pyrene. About 90% of the paired PAH concentrations in laboratory-exposed and field-collected oligochaetes were within a factor of three of one another indicating laboratory results could be extrapolated to the field with a reasonable degree of certainty.

Over the past 10 years, a variety of methods have been described for evaluating the toxicity of sediment-associated contaminants to benthic invertebrates. However, only a limited number of methods are currently available for assessing bioaccumulation of contaminants from field-collected or laboratory-spiked sediments (Ingersoll *et al.* 1995). Standard guides have been published for conducting 28-day bioaccumulation tests with the oligochaete *Lumbriculus variegatus,* including determination of bioaccumulation kinetics for different compound classes (US EPA, 1994; ASTM 1998). *L. variegatus* was selected for use in laboratory bioaccumulation testing in the present study of sediments collected from Upper Mississippi River (UMR) for the following reasons: (1) ease of culture and handling, (2) known chemical exposure history, (3) adequate tissue mass for chemical analyses, (4) tolerance of a wide range of sediment physiochemical characteristics, (5) low sensitivity to contaminants associated with sediment, and (6) amenability to long-term exposures without feeding. Other organisms do not meet many of these selection criteria including mollusks (valve closure), midges (short life cycle), mayflies (difficult to culture), amphipods (small tissue mass, too sensitive), cladocerans and fish (not in direct contact with sediment) (US EPA 1994; ASTM 1998).

Several investigators have conducted bioaccumulation studies with *L. variegatus* using either field-collected or laboratory-spiked sediments (Schuytema *et al.* 1988; Nebeker *et al.* 1989; Ankley *et al.* 1991; Call *et al.* 1991; Carlson *et al.* 1991; Ankley *et al.* 1993; Kukkonen and Landrum 1994). However, only one previous study has compared results of laboratory bioaccumulation studies conducted with *L. variegatus* to residues from synoptically collected field populations of oligochaetes (Ankley *et al.* 1992). Good agreement was reported in this previous study between concentrations of polychlorinated biphenyls in the laboratory and field organisms, particularly for PCBs with K_{ow} values <7. This suggests that laboratory exposures longer than 28 days may be required to reach equilibrium for super-hydrophobic chemicals (Ankley *et al.* 1992).

* *Present address:* Robert S. Kerr Environmental Research Laboratory, United States Environmental Protection Agency, 919 Kerr Research Drive, Ada, Oklahoma 74820, USA

Correspondence to: E. L. Brunson

The United States Geological Survey (USGS) has been monitoring the Upper Mississippi River since 1987 to document the fate and transport of contaminated sediments (Moody and Meade 1995; Moody 1996). Concern with the redistribution of these contaminated sediments arose after the flood of 1993. This project is designed to evaluate the status of sediments in the UMR and is one article in a series designed to assess the extent of sediment contamination in navigational pools of the river. The overall project consists of the following studies: (1) measuring concentrations of contaminants in sediments of the UMR (Moody 1996); (2) toxicity testing with sediments collected from the river (Kemble *et al.* 1998; Winger and Lasier 1998); (3) analysis of benthic community structure (Canfield *et al.* 1998); and (4) bioaccumulation of sediment associated contaminants (the present study). The present study had two objectives: (1) to assess the bioaccumulation of contaminants in sediments collected from the UMR in the laboratory using the oligochaete *L. variegatus,* and (2) to compare bioaccumulation in these laboratory-exposed oligochaetes to synoptically collected field populations of oligochaetes from the UMR.

Materials and Methods

Sample Collection

Sediment samples and native oligochaetes were collected from 23 navigational pools along the UMR and from the Saint Croix River ("C" samples described in Kemble *et al.* 1998 and US EPA 1997). These sampling stations were selected based on the potential of the presence of oligochaetes or fine grained sediment (see Figure 1 in Kemble *et al.* 1998 for a map of these sampling locations on the river). For each of these C samples, 35 to 80 L of sediment (six to 25 grabs) were collected with a stainless steel Ponar grab sampler (Wildlife Supply Company, Saginaw, MI). All grabs from a station within a pool were collected within a 5-m radius and combined in a 114-L high-density polyethylene (HDPE) container. The composited sample was homogenized on the research ship Acadiana using an electric drill and a stainless steel auger. Once homogenized, the following subsamples of sediment were removed: (1) three separate 250-ml subsamples for organic chemistry, metals/acid-volatile sulfides, and total organic carbon/particle size (see Kemble *et al.* 1998 and US EPA 1997 for results of these analyses); (2) one 2-L subsample for benthic invertebrates (Canfield *et al.* 1998); and (3) one 10-L subsample for laboratory toxicity testing (Kemble *et al.* 1998) and bioaccumulation testing in the present study. Sediment samples were stored at 4°C until used in laboratory exposures or until physical and chemical analyses were performed.

The remainder of the composited C sample of sediment was rinsed on ship through a Wildco wash bucket (US Standard sieve size #30, 600-μm opening). The material captured by the wash bucket was transferred to an HDPE tub along with river water. After all the sediment was sieved, native oligochaetes were isolated from the detritus. The oligochaetes isolated from each sample were placed in a HDPE jar containing aerated river water and were held for 24 h to depurate gut contents (US EPA 1994). After the 24-h elimination period, dead oligochaetes were discarded. The remaining oligochaetes were rinsed, blotted dry, weighed, transferred to clean glass jars, and frozen at −22°C until analyzed for chemical contaminants. Weights of native oligochaetes selected for analysis ranged from 0.34 g (Pool 4) to 9.8 g (Pool 9).

Laboratory Testing

L. variegatus were exposed in 28-day sediment exposures following methods described in US EPA (1994) and ASTM (1998). Sediment from 13 of the 23 sampled pools were selected for evaluation in these laboratory exposures. Samples were chosen for testing on the basis of sufficient mass of field-collected oligochaetes for chemical analyses or previously documented presence of PCBs (Pool 4 in lower Lake Pepin; *e.g.,* Rostad *et al.* 1986). Concentrations of contaminants were low in these 23 sediment samples (Kemble *et al.* 1998); therefore, it is unlikely that the distribution of oligochaetes in these samples was substantially influenced by contaminants (Canfield *et al.* 1998). Oligochaetes used in laboratory testing were mass cultured in 75-L glass aquarium containing 50 L of well water (hardness 290 mg/L as $CaCO_3$, alkalinity 255 mg/L as $CaCO_3$, pH 7.8; US EPA 1994, ASTM 1998). Each culture aquarium received about 27 volume additions (about 1.5 L/minute) of well water daily. The culture water was aerated and maintained at 23°C. Presoaked, shredded brown paper towels were used as substrate. Cultures of oligochaetes were fed Tetramin flake fish food twice weekly *ad libitum.*

Exposures of oligochaetes were conducted in 4-L glass Pyrex beakers containing 1 L of sediment and 3 L of overlying water. Four replicate chambers were tested for each of the 13 sediment samples evaluated. Reconstituted water was used as source of overlying water (hardness 90 to 96 mg/L as $CaCO_3$, alkalinity 60 to 70 mg/L as $CaCO_3$; US EPA 1994). Each beaker was calibrated to 4 L using a glass standpipe that exited through the beaker wall and was held in place with a silicon stopper. Beakers received two volume additions (6 L ± 10%) of overlying water per day. Water was delivered using a modified Mount and Brungs diluter system that was designed to deliver 1 L/cycle (Ingersoll and Nelson 1990). An in-line flow splitter was attached to each delivery line to split the water flow evenly to each of four beakers. These splitters were constructed of quarter-inch PVC pipe with four silicone stoppers and 14-gauge stainless steel hypodermic needles with the points and connector ends cut off the needles (Figure 1). Glass stands were used to support the splitters keeping them level to maintain a constant volume delivery to each beaker (±5%). Beakers were held in a temperature-controlled waterbath (23 ± 1°C) on a 16:8 light:dark photoperiod at about 500 lux. Oligochaetes were not fed during the sediment exposure.

Sediment and overlying water were placed in the beakers the day before adding organisms (Day −1). Sediments were first homogenized with a hand-held electric drill and stainless steel auger before being placed into the beakers (Kemble *et al.* 1998). One liter of sediment was transferred into each beaker using a plastic spoon. Overlying water was poured into the beakers through a piece of fine-mesh Nitex® material to minimize suspension of the sediment. Delivery of overlying water was started after beakers were placed in the waterbath.

Twenty-four hours before stocking the test (Day −1) oligochaetes were removed from the culture with a fine-mesh nylon aquarium net, placed in beakers containing well water, and rinsed to remove excess toweling and debris. Beakers containing the oligochaetes were then placed in a waterbath and aerated. With substrate absent, the *L. variegatus* formed tight clumps in the beakers, which was helpful during transfer of organisms into the beakers containing sediment.

Oligochaetes were acclimated to the test water by removing half of the water in each beaker and replacing it with temperature-acclimated test water. Two hours later this process was repeated. After another 2 h, the *L. variegatus* were combined into a glass pan and rinsed with test water to break up the masses of worms and remove any remaining debris. With the mass of worms disturbed, oligochaetes were grouped together with a stainless steel dental pick and allowed to form small clumps of about 1 g. The clumps of oligochaetes were removed from the pan with the dental pick, touched against the rim of the pan to remove excess water, and placed on a tared weigh boat. About 2.6 g of unblotted oligochaetes were transferred to each beaker containing sediment and overlying water. Using this approach, the 2.6 g of

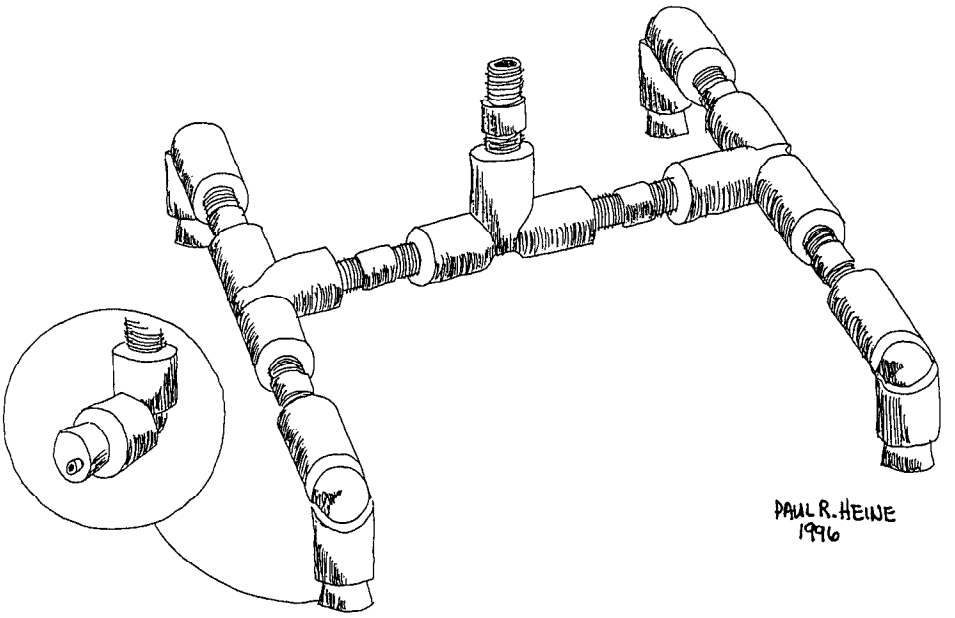

Fig. 1. Diagram of in-line flow splitter used to deliver overlying water in the sediment exposures of *Lumbriculus variegatus* in the laboratory

unblotted oligochaetes represents about 2 g of blotted oligochaetes or about 200 organisms.

General conditions of the exposure system and behavior of the oligochaetes were evaluated daily. Dissolved oxygen and conductivity of the overlying water were measured weekly in all beakers. Total hardness (as $CaCO_3$), pH, alkalinity (as $CaCO_3$), and total ammonia of overlying water were measured at the beginning and end of the test. Overlying water pH, alkalinity, total hardness, conductivity and total ammonia measurements were similar among all stations and inflowing test water (US EPA 1997). Dissolved oxygen measurements were at or above acceptable levels (>40% of saturation; ASTM 1998) in all treatments throughout the study (US EPA 1997). Ranges of mean water quality for each parameter were as follows: pH 7.7–7.9; alkalinity as $CaCO_3$ 61–67 mg/L; total hardness as $CaCO_3$ 104–110 mg/L; conductivity 342–350 µS at 25°C; total ammonia 0.1–0.4 mg/L; and calculated unionized ammonia 0.0028–0.0094 mg/L (US EPA 1997).

On day 28 of the exposure, *L. variegatus* were isolated from each beaker by washing the sediment through No. 18 (1.0-mm opening) followed by No. 50 (300-µm opening) US standard stainless steel sieves. The contents of each sieve was rinsed into several clear glass pans and all oligochaetes were removed. *L. variegatus* were separated from native oligochaetes based on behavior (native oligochaetes tended to form a tight, spring-like coil, whereas *L. variegatus* would not) (US EPA 1994). Once isolated, all *L. variegatus* from a beaker were cleaned of any remaining debris and held for 24 h in 1-L water-only beakers to allow them to clear their gut contents (US EPA 1994). The *L. variegatus* were then isolated, cleaned of any remaining debris, and transferred to a tared weigh boat. Samples were then blotted, weighed, placed in glass jars, and stored at −22°C pending chemical analysis for contaminants. Sample weights of replicate laboratory-exposed oligochaetes from each beaker ranged from 1.3 to 3.0 g after the 28-day exposure to sediment.

Chemical Analyses

Chemical characterizations of sediments included: organochlorine pesticides (OCs), polychlorinated biphenyls (PCBs), select aliphatic and polynuclear aromatic hydrocarbons (PAHs), and total organic carbon. See Kemble *et al.* (1998) for a description of these methods and for the results of these sediment analyses. Kemble *et al.* (1998) also describes results of physical characterizations and analyses of metals in these sediment samples.

Concentrations of metals and organochlorine compounds in sediment samples were low (US EPA 1997; Kemble *et al.* 1998). Therefore, replicate tissue samples from the laboratory exposures were combined for organochlorine pesticide/PCB analyses and metals were not analyzed because of limited mass of samples. Tissues were analyzed by Geochemical and Environmental Research Group at Texas A&M University, College Station, Texas for the following: (1) OCs, (2) PCBs, (3) select PAHs, and (4) percent lipid. Before analysis, tissue samples were homogenized and extracted using a Teckmar Tissumizer, sodium sulfate, and methylene chloride (MacLeod *et al.* 1985; Wade *et al.* 1988; Brooks *et al.* 1989). Tissue extracts were split into two fractions: one fraction was used to measure percent lipid and the second fraction was used for measuring PAHs, OCs, and PCBs. Extracts for chemical analyses were purified using absorption chromatography to isolate the aliphatic fraction and the PAH/OC/PCB fraction. Lipid interference in the PAH/OC/PCB fraction was eliminated with further purification using high pressure liquid chromatography (HPLC). The quantitative analyses were performed by capillary gas chromatography (CGC) with electron capture detector for OCs and PCBs and a mass spectrometer detector in the selected ion monitoring mode for PAHs (Wade *et al.* 1988).

Percent lipids were calculated on a wet-weight basis. A 20-ml aliquot of the total extract volume of about 300 ml was filtered, concentrated to 1 ml, and weighed. A 100-µl subsample was then removed, evaporated to dryness, and weighed. Percent lipid was calculated using the weight of the dried subsample and the concentrated sample. Raw data for tissue and sediment analyses are presented in US EPA (1997) and are also available through the Internet at our home page (http://www.ecrc.cr.usgs.gov/pubs/umr.html).

Average percent spike recovery for 22 OCs was 88% (n = 4). Beta BHC had the smallest average spike recovery (53%) while oxychlordane had the greatest average spike recovery (104%). Individual OC concentrations were often below minimum detectable limits, so duplicate analyses were evaluated only for total PCBs. The average duplicate coefficient of variation was 26% (range 0.7–61%, n = 4). Average percent spike recovery for PAH compounds was 96% (25 compounds, n = 4). Indeno(c,d)pyrene had the smallest average per-

cent recovery (81%) while 1-methylnaphthalene had the greatest average percent recovery (110%). The average duplicate coefficient of variation was 21% (34 possible compounds, n = 1 to 4). Average duplicate coefficient of variation ranged from 1% for C1-phenanthracene to 79% for benzo(a)pyrene.

In addition to the laboratory-exposed and field-collected oligochaetes, three samples of oligochaetes from laboratory cultures were collected at the beginning of the exposure for analysis contaminants. Two of the three samples had detectable concentrations of PAHs and total PCBs however, the concentrations were generally less than those of oligochaetes exposed to or collected from the sediments from the UMR. For some unexplained reason, concentrations of total PCB (1.3 μg/g wet weight) and some PAHs (up to 0.25 μg/g wet weight) detected in one of those three samples was similar to oligochaetes exposed during the test.

Results and Discussion

General Trends

Individual organochlorine pesticides (OCs) were generally below the detection limits (ranging from 0.0007 to 0.0217 μg/g wet weight for both laboratory-exposed and field-collected oligochaetes) (US EPA 1997). Only six of these OC compounds were detected in the 13 field-collected samples of oligochaetes (22 compounds evaluated). The greatest individual OC concentration was 0.009 μg/g (wet weight) for dieldrin in oligochaetes collected from Pool 22. As was the case with the field-collected oligochaetes, tissue concentrations of individual OCs were typically below detection limits in the laboratory-exposed oligochaetes (US EPA 1997). All of the laboratory-exposed oligochaetes had at least one OC concentration above background (Pool 13 and Pool 16; 4,4′-DDE); however, no sample had more than six OCs detected (Pool 11 and 14; gamma-chlordane, alpha-chlordane, aldrin, dieldrin, 4,4′-DDE, 4,4′-DDD). The greatest individual OC concentration was 0.013 μg/g (wet weight) for 4,4′-DDE for the oligochaetes exposed in the laboratory to sediment collected from Pool 4. Also, 4,4′-DDE was the most frequently measured OC (12 samples) in laboratory-exposed oligochaetes with concentrations ranging from 0.0021 to 0.013 μg/g (wet weight).

Total PCBs were the only chlorinated organic compounds detected in all field-collected and laboratory-exposed oligochaetes. Concentrations ranged from 0.045 μg/g (wet weight, Pool 13) to 0.697 μg/g (wet weight, Pool 4). The geometric mean for total PCBs measured in either oligochaetes exposed to the sediment samples in the laboratory or collected from the field was 0.18 μg/g.

Field-collected and laboratory-exposed oligochaete samples were analyzed for 44 PAH congeners. Field-collected oligochaetes from Pool 4 had the fewest number of PAHs detected (14) while Pool 19 had the most number of PAHs detected (36). Only 16 PAH congeners (about 40% of those analyzed for) were detected in samples from seven of the 13 pools for both the field-collected and laboratory-exposed oligochaetes (detection limits of 0.0217–0.0024 μg/g; selection criteria: two of the four replicates of tissue samples from the laboratory exposures needed to have a detected concentration).

Table 1 lists the 16 PAHs measured in tissue samples that met the selection criteria described above. Figures 2 and 3 depict accumulation of these PAHs from laboratory-exposed or field-collected oligochaetes for each UMR pool evaluated. Concentrations of these 16 PAH congeners were converted to molar units, normalized to percent lipid, and summed. Oligochaetes exposed to in the laboratory to sediments from Pool 7 were more contaminated than oligochaetes exposed in the laboratory to sediments from the other pools (Figure 2). Field-collected oligochaetes from Pool 4 were more contaminated than oligochaetes from the other pools (Figure 3). In general, perylene had the highest concentration of any PAH in the laboratory-exposed or field-collected oligochaetes. Perylene concentrations ranged from 0.0561 to 0.5299 μg/g (wet weight) in field-collected oligochaetes and from 0.0516 to 0.8397 μg/g (wet weight) in the laboratory-exposed oligochaetes.

Table 1. List of polycyclic aromatic hydrocarbons (PAHs) that met our selection criteria for laboratory to field comparisons of tissue concentrations and their associated molecular weight and log K_{ow}[a]

Chemical No.		Molecular Weight	Log K_{ow}	Plot Pattern
Low Molecular-Weight PAHs				
1	Naphthalene	128.17	3.35	A
2	1-methylnaphthalene	142.20	3.87	A
3	2-methylnaphthalene	142.20	4.00	A
4	Biphenyl	154.21	3.90	B
5	2,6-dimethylnaphthalene	156.23	4.31	A
6	Fluorene	166.22	4.38	B
7	1,6,7-trimethylnaphthalene	170.25	4.70	A
8	Phenanthrene	178.23	4.57	C
9	1-methylphenanthrene	192.26	5.14	B
High Molecular-Weight PAHs				
10	Pyrene	202.26	5.18	B
11	Fluoranthene	202.26	5.22	B
12	Chrysene	228.29	5.86	B
13	Benzo(a)anthracene	228.29	5.91	C
14	Benzo(b,k)fluoranthene	252.32	6.20	C
15	Perylene	252.32	6.25	C
16	Benzo(e)pyrene	252.32	6.44	B

[a] An "A" indicates compounds where the concentrations in the field > laboratory, "B" indicates compounds where concentrations the laboratory were similar to field, and "C" indicates compounds where the concentrations in the laboratory > field

Samples of sediments (Kemble *et al.* 1998; US EPA 1997) and oligochaetes (Figure 2 and 3) from the UMR are relatively uncontaminated compared to other locations we have previously evaluated using sediment toxicity tests (Ingersoll *et al.* 1996) or bioaccumulation tests (studies of the Little Scioto River in Ohio and Huntsville River in Alabama; Brunson *et al.* 1993). In the present study, tissue concentrations of PAHs were generally greatest in field-collected oligochaetes from Pool 4 (Figure 3). Two low molecular weight (LMW) PAHs (naphthalene and phenanthrene) and two high molecular weight (HMW) PAHs (pyrene and chrysene) were generally the PAHs of highest concentration in field-collected oligochaetes from Pool 4. Concentrations for these compounds in sediment were relatively low (1.4 to seven times below reported effect range medians (ERMs) for these compounds; Ingersoll *et al.* 1996, Kemble *et al.* 1998). Tissue concentrations of PAHs in oligochaetes collected from UMR Pool 4 were more than two orders of magnitude less than tissue concentrations of oligochaetes exposed to sediments from the Little Scioto River in Ohio. Collectively, this information indicates that sediment and biota from the UMR is relatively uncontaminated compared to other contaminated sites previously evaluated by our laboratory.

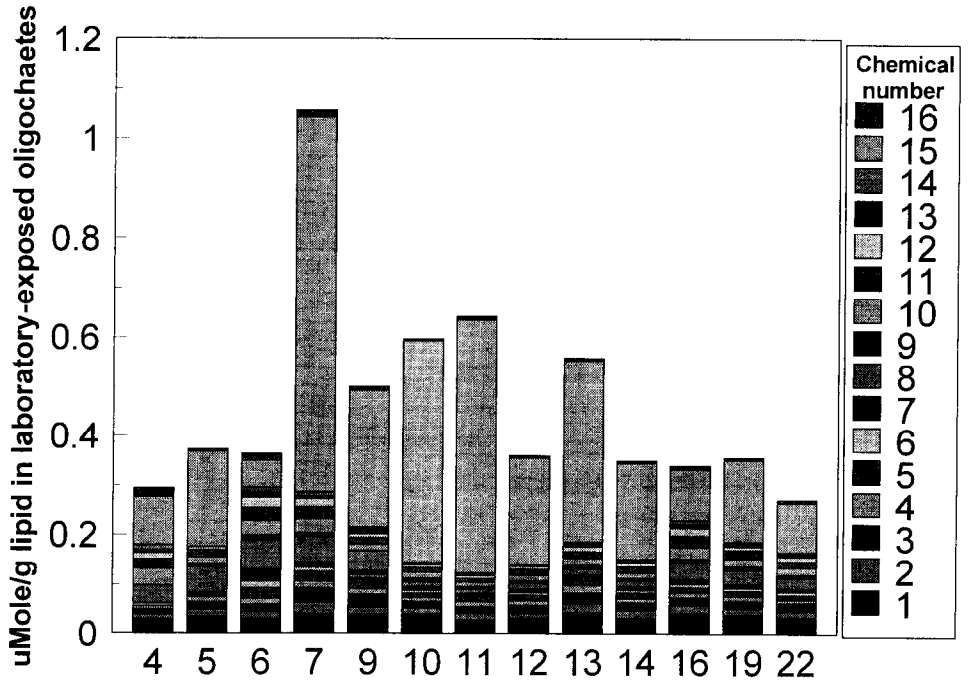

Fig. 2. Total accumulation of polycyclic aromatic hydrocarbons (PAHs; μMole/g lipid) by *Lumbriculus variegatus* exposed in the laboratory to sediments from the Upper Mississippi River. Chemical numbers correspond to the following compounds: (1) naphthalene, (2) 1-methylnaphthalene, (3) 2-methylnaphthalene, (4) biphenyl, (5) 2,6-dimethylnaphthalene, (6) fluorene, (7) 1,6,7-trimethylnaphthalene, (8) phenanthrene, (9) 1-methylphenanthrene, (10) pyrene, (11) fluoranthene, (12) chrysene, (13) benzo(a)anthracene, (14) benzo(b,k)fluoranthene, (15) perylene, and (16) benzo(e)pyrene

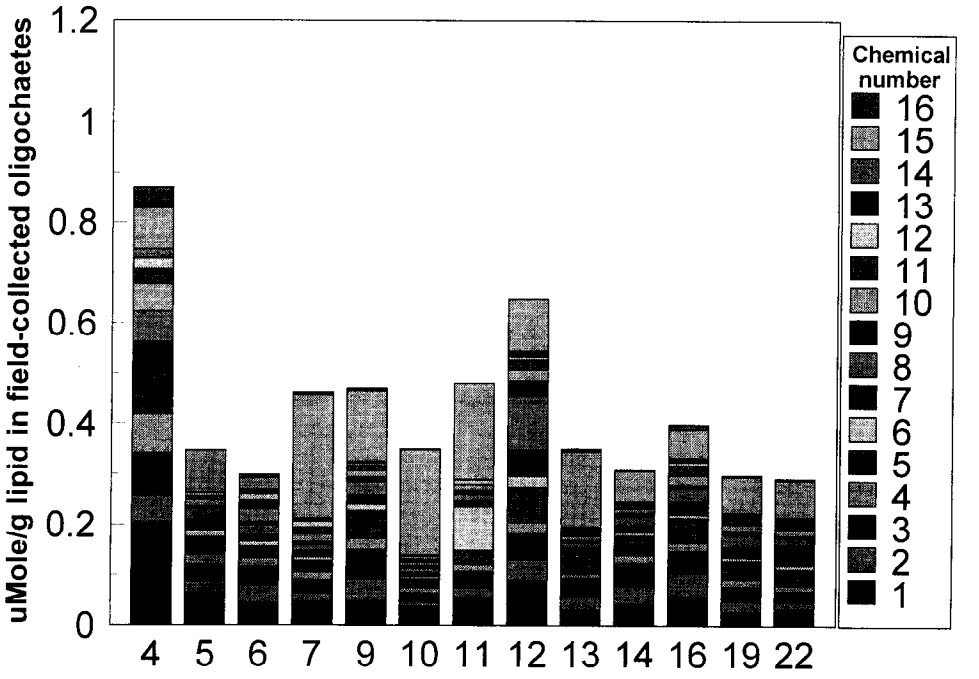

Fig. 3. Total accumulation of polycyclic aromatic hydrocarbons (PAHs; μMole/g lipid) by oligochaetes collected from select pools of the Upper Mississippi River. Chemical numbers for individual compounds correspond to those listed for Figure 2

Detection of Compounds in Tissue vs. Sediment

Detection limits for compounds in sediment (Kemble *et al.* 1998) and tissue are usually different, which creates difficulties in interpreting bioaccumulation data in relatively uncontaminated sediments. In the UMR, concentrations of PAHs and PCBs were detected in both sediments and laboratory-exposed oligochaetes 79% of the time, and were detected in both sediment and field-collected oligochaetes 58% of the time. Concentrations of PAHs and PCBs were not detected in the

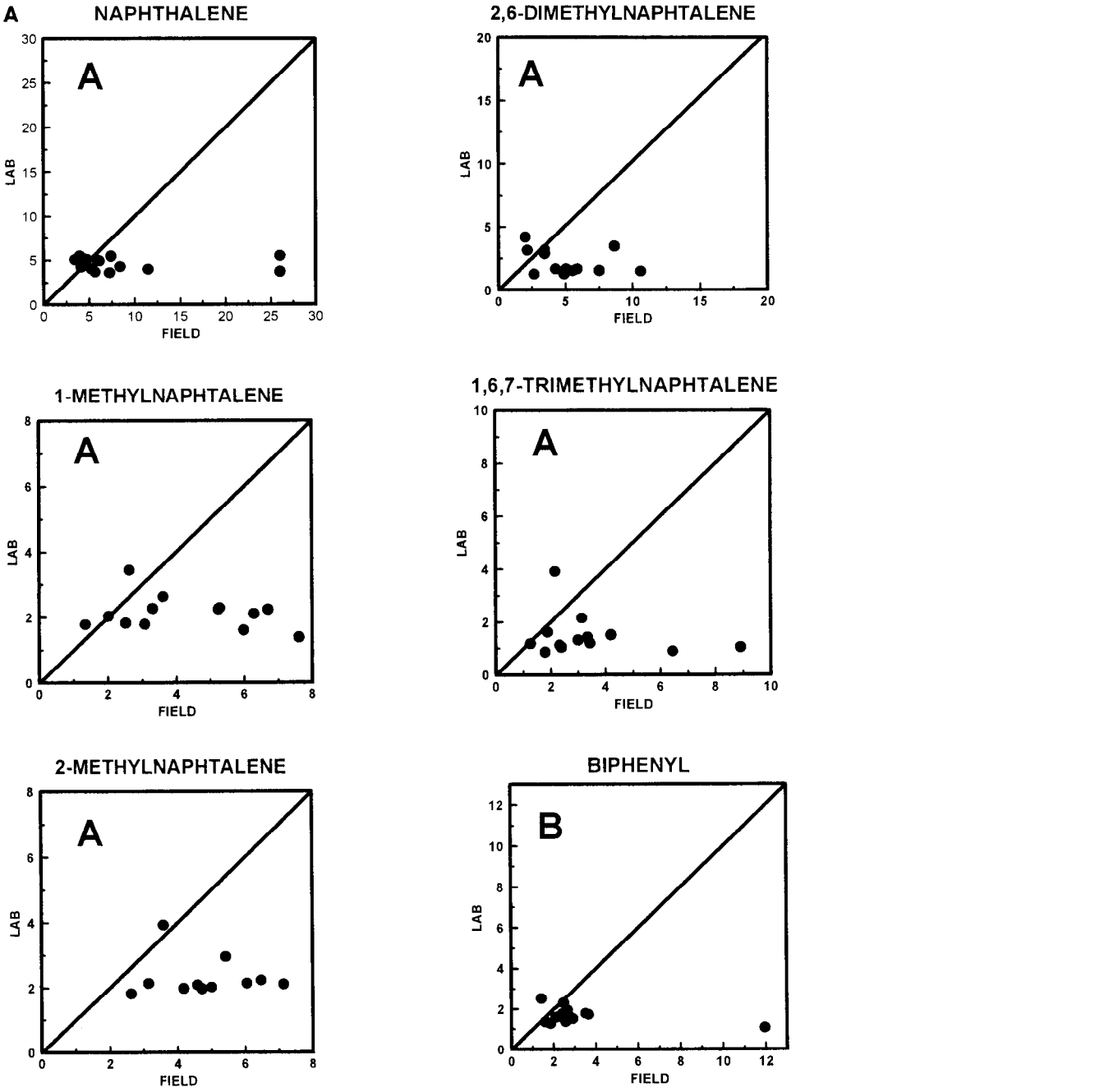

Fig. 4. Comparison of tissue concentrations in laboratory-exposed *Lumbriculus variegatus* compared to field-collected oligochaetes. An "A" indicates concentrations where the field > lab, "B" indicates concentrations where laboratory was similar to the field, and "C" indicates concentrations where the laboratory > field

sediments and were detected in laboratory-exposed oligochaetes in 17% of the samples and were not detected in sediment and were detected in field-collected oligochaetes in 41% of the samples. Concentrations of PAHs and PCBs were detected in sediment samples but not detected in laboratory-exposed oligochaetes in 3% of the samples and were detected in sediment samples but not detected in field-collected oligochaetes in 1% of the samples. In the present study, the detection limits for measuring organic contaminants in sediments and tissues met recommended guidelines (US EPA 1984); however, these detection limits for sediments should have been lower in order to better represent bioavailable compounds in organisms inhabiting the UMR.

Laboratory to Field Comparisons

Tissue concentrations of naphthalenes were generally higher in field-collected oligochaetes than in laboratory exposed oligochaetes (Figure 4). Naphthalenes are LMW PAHs with log K_{ow} values less than 4.5. PAHs with similar concentrations in both the laboratory-exposed and field-collected oligochaetes in-

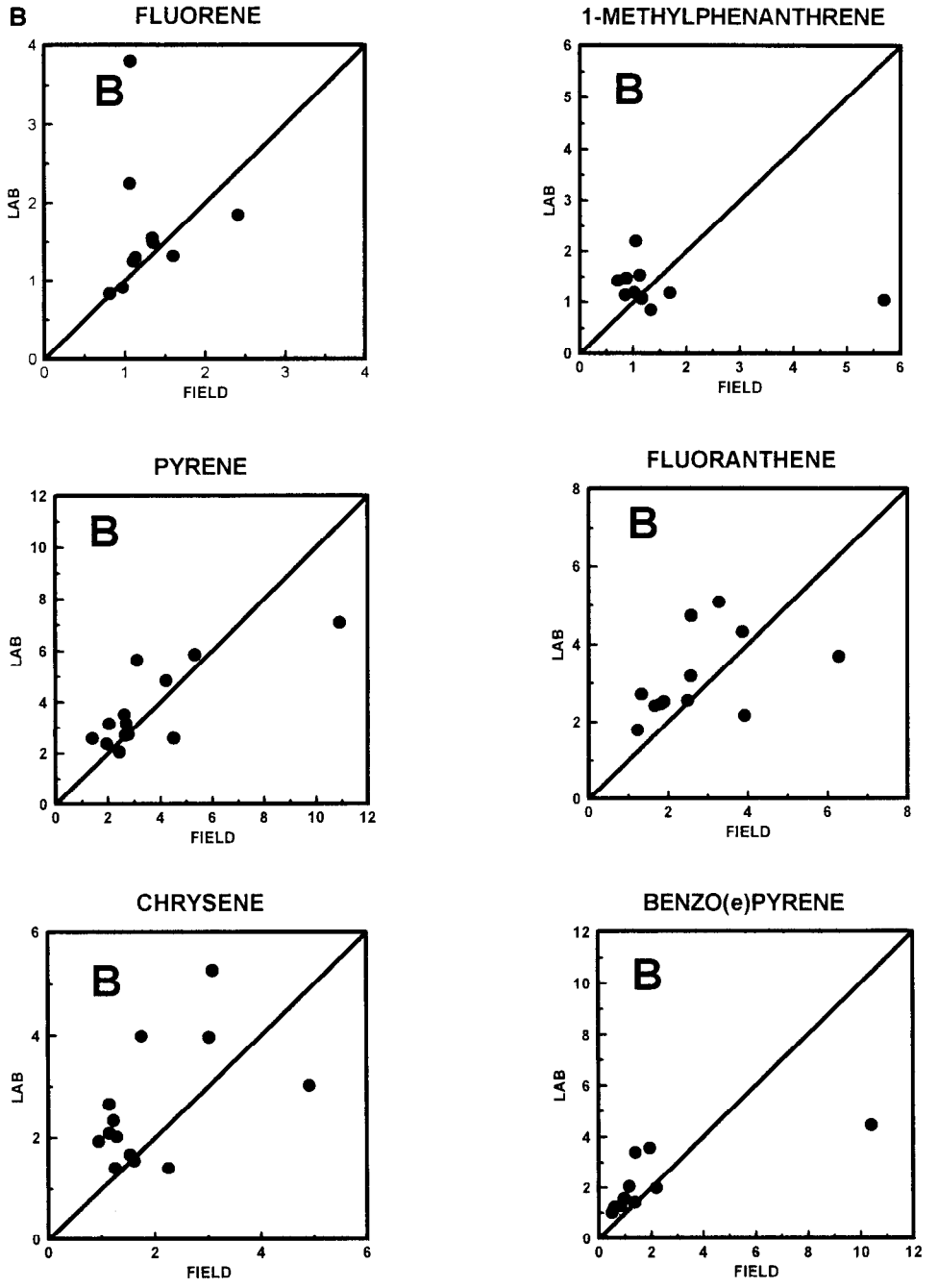

Fig. 4. Continued

cluded a similar number of HMW and LMW compounds (biphenyl, fluorene, 1-methylphenanthrene, pyrene, fluoranthene, chrysene, and benzo(e)pyrene). Most of these compounds are intermediate in molecular weight and log K_{ow} (except for benzo(e)pyrene which was one of the highest molecular weight and log K_{ow} of all compounds included in Figure 4). PAHs typically higher in the laboratory-exposed compared to the field-collected oligochaetes were primarily HMW compounds (benzo(a)anthracene, benzo(b,k)fluoranthene, and perylene) with a log K_{ow} of greater than 5.1 (Figure 4 and Table 1).

A positive correlation was observed between lipid-normalized concentrations of PAHs detected in laboratory-exposed and field-collected oligochaetes across all sampling locations (Spearman rank correlation (r) = 0.51, p = 0.0001, n = 185) (SAS 1994). Rank correlations for concentrations of individual compounds between laboratory-exposed and field-collected oligochaetes were strongest for benzo(e)pyrene (r = 0.88, p = 0.0008, n = 10), perylene (r = 0.87, p = 0.0001, n = 13), benzo(b,k)fluoranthene (r = 0.69, p = 0.058, n = 8), and pyrene (r = 0.71, p = 0.061, n = 13).

The ratio of tissue concentrations of PAHs in laboratory-exposed oligochaetes to concentrations in field-collected oligochaetes were generally similar (Figure 5). About 90% of the corresponding concentrations were within a factor of three between the laboratory-exposed and field-collected oligo-

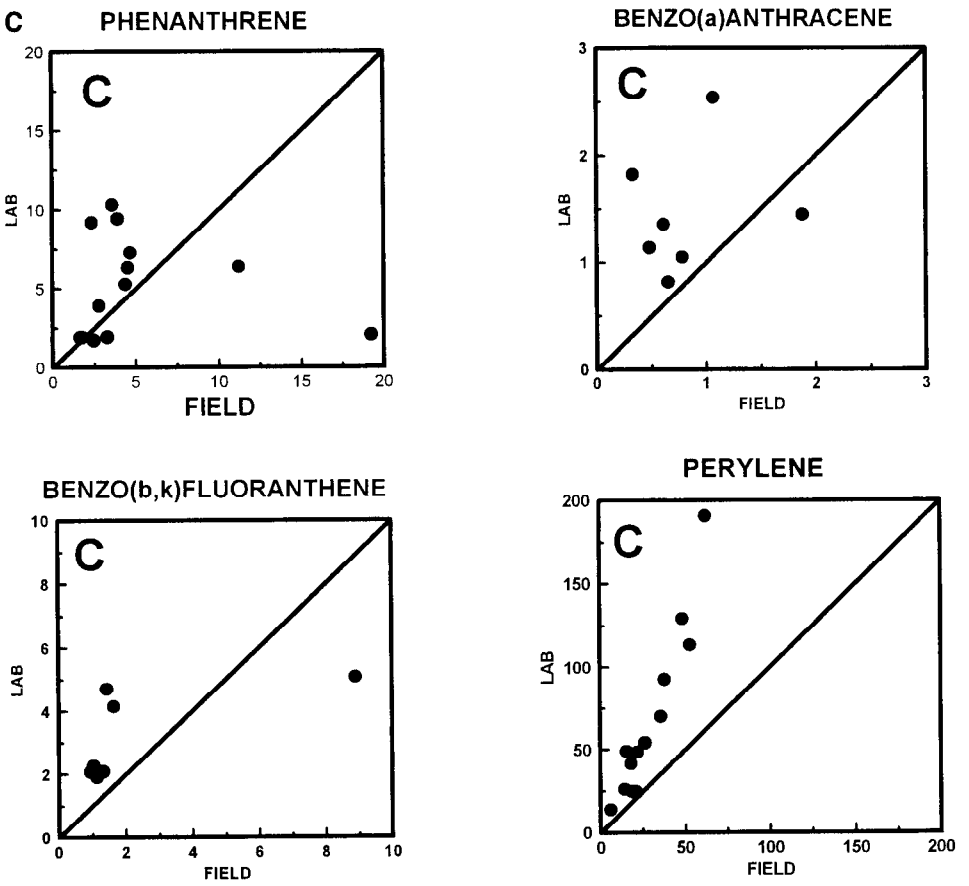

Fig. 4. Continued

chaetes (represented by the crosshatched region in Figure 5). However, there appears to be a shift from concentrations in the field > lab to concentrations in the lab > field as the molecular weight of PAHs increases. Concentrations that differed by more than a factor of three were primarily LMW PAHs (naphthalene, 1-methylnaphthalene, 2-methylnaphthalene, 2,6-dimethylnaphthalene, fluorene, 1,6,7-trimethylnaphthalene, phenanthrene, and 1-methylphenanthrene) and were usually elevated in the field-collected oligochaetes compared to the laboratory-exposed oligochaetes. Ratios >3 in the laboratory-exposed or field-collected oligochaetes were most frequently associated within a small number of pools (Field > 3× lab in Pools 4, 12, 22; lab > 3× field in Pool 7).

Differences between tissue concentrations in the laboratory-exposed and field-collected oligochaetes may have resulted from relatively water-soluble LMW PAHs being lost during the sampling of sediments. A second possibility for differences between the laboratory and field-exposed may be spatial heterogeneity of contaminants in the sediments in the field. Other possible explanations could include the route of exposure. Exposure to contaminants in the field may occur through sediment, food, and overlying water, while the route of exposure to oligochaetes in the laboratory was sediment. Species-specific differences in exposure between *L. variegatus* and the native oligochaetes may also contribute to the differential accumulation.

Biota-Sediment-Accumulation Factors

Biota-sediment-accumulation factors (BSAFs) (US EPA 1994) were calculated by dividing the lipid-normalized tissue concentrations by the organic-carbon normalized sediment concentrations reported in Kemble *et al.* (1998) and in US EPA (1997). Mean BSAFs for this study were only listed for compounds for which BSAF could be calculated for both laboratory-exposed and field-collected oligochaetes in at least seven of 13 pools (Table 2). Mean BSAFs for laboratory-exposed oligochaetes ranged from 0.97 for benzo(a)anthracene to 5.3 for naphthalene. Mean BSAFs for field-collected oligochaetes ranged from 1.1 for chrysene to 8.8 for naphthalene. Individual BSAFs for naphthalene ranged from 1.6 to 10.1 in laboratory-exposed oligochaetes and from 2.5 to 26.6 in field-collected oligochaetes. BSAFs for pyrene, benzo(a)anthracene, and benzo(b,k) fluoranthene were typically greater than BSAFs reported for marine organisms (Lee 1992). BSAFs were also calculated using PCB homolog data reported in Ankley *et al.* (1992) for laboratory-exposed *L. variegatus* and field-collected oligochaetes (Figure 6). BSAFs were similar between laboratory-exposed and field-collected oligochaetes in both Ankley *et al.* (1992) and in the present study; however, BSAFs in the present study were typically greater (0.97–8.8) than those from Ankley *et al.* (1992) (0.17–2.26).

A theoretical value of 1.7 for BSAFs has been estimated based on partitioning of nonionic organic compounds between

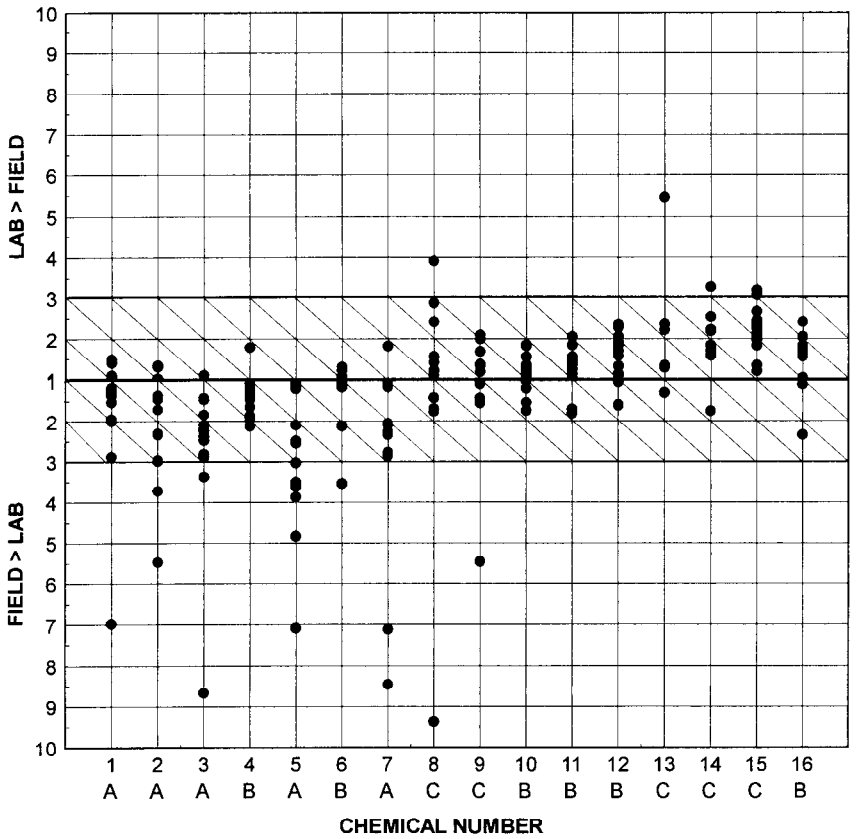

Fig. 5. Ratio of lipid-normalized tissue concentrations in laboratory-exposed or field-collected oligochaetes for select PAHs. See the legend to Figure 2 for a listing of the specific compounds by chemical number. An "A" indicates concentrations where the field > laboratory, "B" indicates concentrations where the laboratory was similar to field, and "C" indicates concentrations where the laboratory > field. Compounds are plotted in order of molecular weight with molecular weight increasing from left to right. If the laboratory concentration of a compound for a pool is higher than the corresponding field concentration, then the laboratory/field ratio is plotted on the upper half of the plot. If the field concentration of a compound for a pool is higher than the corresponding laboratory concentration, then the field/laboratory ratio is plotted on the lower half of the plot (see US EPA 1997 for a listing of ratio values)

Table 2. Biota-sediment-accumulation factors (BSAFs) reported by Lee (1992) and in the present study[a]

Compound	Lee (1992)	Range	Lab	Range, SEM, n	Field	Range, SEM, n
Naphthalene	—	—	5.3	1.6–10.1, 0.54, 13	8.8	2.5–26.6, 1.82, 13
2-Methylnaphthalene	—	—	2.6	0.9–5.1, 0.31, 12	6.7	2.2–12.2, 0.99, 12
Pyrene	0.4	0.18–0.5	2.3	0.8–3.9, 0.32, 12	2.2	0.7–5.6, 0.43, 12
Fluoranthene	—	—	1.8	0.9–3.9, 0.27, 12	1.6	0.6–4.9, 0.34, 12
Chrysene	—	—	1.5	0.7–2.4, 0.15, 12	1.1	0.3–2.0, 0.15, 12
Benzo(a)anthracene	0.4	0.2–0.6	0.97	0.4–2.5, 0.30, 7	—	—
Benzo(b,k)fluoranthene	0.4	0.2–1.0	—	—	—	—
Perylene	—	—	2.2	0.5–4.7, 0.35, 13	1.0	0.3–1.9, 0.15, 13

[a] The means BSAFs for the present study are listed where there was matching detection of a particular compound in both sediment and tissue in at least seven of 13 pools for laboratory-exposed (lab) or field-collected (field) oligochaetes. The range, standard error of the mean, and n for BSAFs in the present study are reported as means for samples of individual pools of the upper Mississippi River

sediment carbon and tissue lipids (McFarland and Clarke 1986). A BSAF of less than 1.7 indicates less partitioning into lipids than predicted and a value greater than 1.7 indicates more uptake than can be explained by partitioning theory alone (Lee 1992). The majority of the BSAFs in Table 2 were within a range of about 1.0–2.6, suggesting the theoretical BSAF value of 1.7 could be used to predict these mean BSAFs with a reasonable degree of certainty. However, mean BSAFs for naphthalene (8.8) and 2-methylnaphthalene (6.7) in the field-collected oligochaetes were elevated relative to a theoretical BSAF of 1.7. Moreover, BSAFs for individual pools were as high as 10.1 for laboratory-exposed oligochaetes and 26.6 for field-collected oligochaetes. The higher BSAFs in the field-collected oligochaetes may be the result of: (1) exposure to contaminants in the overlying water; (2) spatial differences in sediment contamination (*i.e.*, sediments were not sampled from

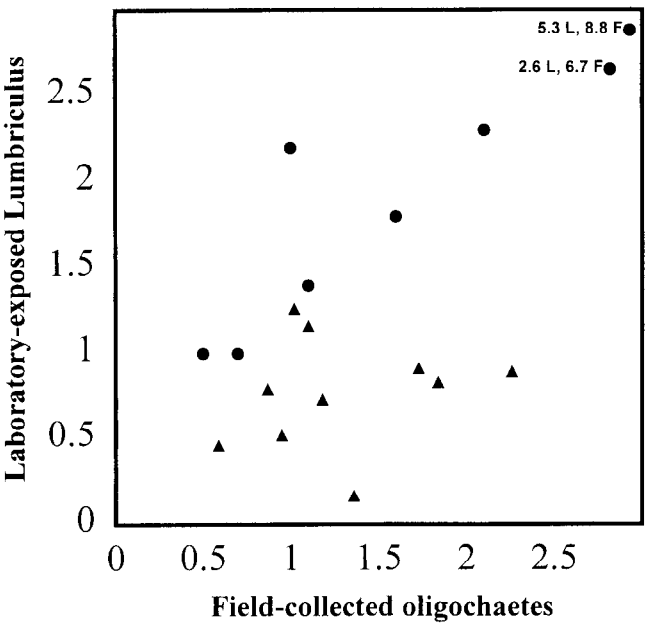

Fig. 6. Biota-sediment-accumulation factors (BSAFs) for laboratory-exposed *Lumbriculus variegatus* and field-collected oligochaetes for PAHs in the present study (circles) and calculated from PCB homolog data reported in Ankley *et al.* (1992, triangles). Two data points (laboratory 5.3, field 8.8 and laboratory 2.6, field 6.7) are not plotted on the same scale

a depth representative of the habitat of the oligochaetes in the field); (3) increased error in chemical analyses due to low concentrations of compounds in sediments or tissues; or (4) taxonomic-specific differences in exposure. BSAFs substantially different from the theoretical value of 1.7 may also result when the system has not reached steady state (*i.e.,* depletion or release of contaminants in pore water in the field). Higher BSAFs in laboratory-exposed oligochaetes may also have resulted from error in chemical analyses due to low concentrations of compounds in sediment or tissue or from disturbance of sediment resulting in a change in equilibrium of compounds in the sediment samples evaluated in the laboratory.

Summary

Contaminant concentrations were relatively low in sediments and tissues collected from the 13 UMR pools evaluated. Only PAHs and total PCBs were frequently measured above detection limits. Previous studies conducted by our laboratory with highly contaminated sediments resulted in accumulation of PAHs up to 1,000 times greater than tissue concentrations observed in the present study. Concentrations of PAHs in laboratory-exposed and field-collected oligochaetes were generally similar. About 90% of the paired PAH concentrations in laboratory-exposed and field-collected oligochaetes were within a factor of three of one another. With the detection limits used to analyze samples in the present study, contaminants were detected in tissue samples more often than in the associated sediment samples. These results indicate detection limits should have been lower in order to better represent the bioavailability of sediment-associated compounds to organisms inhabiting the UMR.

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